Analysis of Fissile Materials by High-Energy Neutron-Induced Fission Decay Gamma Rays[a]

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Experiments were conducted at the Budapest Research Reactor using both cold and thermal guided neutron beams. The thermal beam had a thermal equivalent neutron flux of 2×10^6 cm^{-2} s^{-1}. The cold beam has a thermal equivalent neutron flux of 5×10^7 cm^{-2} s^{-1}. The gamma rays were detected with a Compton suppressed HPGe detector. A beam chopper was also used in some of the measurements and frequency of the chopper was set to 25 Hz. Each cycle was divided into 20 ms activation and counting intervals and spectra were recorded only during the middle 16 ms of each interval. Targets of U_3O_8 with ^{235}U enrichments of natural, 19.1%, 36%, and ≈95% were irradiated in the thermal and cold neutron beams. Gamma-ray spectra were analyzed using the Hypermet PC program. To obtain emission rates, the intensities were corrected for the counting efficiency and saturation of the activity. Decay spectra were corrected for their different dead times, and for activation and counting time. Absolute gamma-ray cross sections were determined by comparison with the standard ^{1}H(2223 keV) gamma ray cross section, 0.3326 b, for targets of $UO_2(NO_3)_2\cdot6H_2O$ and $UO_2(CH_3COO)_2\cdot6H_2O$ with well known stoichiometry.

The Berkeley D+D neutron generator utilizes RF-induction discharge to generate deuterium plasma. The neutron generator is based on a co-axial design, which maximizes the target area in the compact outer dimensions of the generator and enables operation at high beam power, thus yielding high neutron fluxes. The generator was typically operated at an acceleration voltage of 120 kV and 50 mA deuterium beam current. This beam power yielded a ≈2.5 MeV neutron output of 5×10^9 n/s. Gamma rays were detected with a 20% efficient, relative to 3"×3" NaI, HPGe detector. The efficiency was calibrated with a certified multinuclide calibration source from Isotope Product Laboratories. Spontaneous fission neutrons were detected with a ^{3}He neutron detector that also serves as a flux monitor for the neutron generator. A 0.89 kg target of uranium, depleted to 0.1% in ^{235}U, was counted to obtain the natural ^{238}U gamma ray spectrum. It was then irradiated next to the neutron generator for 10 minutes, manually removed from the irradiation chamber, and counted for 6 minutes, starting 1 minute after bombardment. A larger, 14.65 kg, sample of the same depleted uranium was neutron counted with the ^{3}He neutron detector surrounded by a polyethylene moderator.

The prompt and delayed gamma ray spectra produced following the irradiation of uranium enriched to ≈95% in ^{235}U is shown in Figure 1. A ^{238}U decay spectrum, counted for 60 minutes, is inset in Figure 1. In Table 1 the measured fission product gamma ray yields for ^{90}Rb ($t_{1/2}$=258 s), $^{90\text{-}m}$Rb ($t_{1/2}$=158 s), and ^{95}Y($t_{1/2}$=10.3 min) from ^{235}U (E_n=thermal) and ^{238}U (E_n=2.5 MeV) are shown. They are compared with tabulated fission product yields from ENDF-349 for ^{235}U, ^{238}U, and ^{239}Pu where the gamma-ray intensities are inferred using the tabulated yields and the decay normalizations from the Table of Isotopes. Significant differences are observed in the ratios of experimental values for ^{235}U, ^{238}U, and the compiled values for ^{239}Pu suggesting that a combined analysis with thermal and fast neutrons can determine uranium enrichments and the relative abundance of all fissile material. The ^{235}U experimental and ENDF-349 thermal neutron induced fission yields agree well, but the ^{238}U experimental values agree poorly with compilation suggesting possible problems with the database.

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Table 1. Comparison of measured and semi-empirical fission product gamma ray yields.

Fission Product	Eg (keV)	Relative Fission Yields: Experiment ^{235}U(cold)	^{238}U(2.5 MeV)	ENDF-349 Thermal	Fast	^{239}Pu (thermal)
^{90m}Rb(258 s)	3317.00(17)	77	145	70	44	33
^{95}Y(10.3 m)	3575.84(20)	147	325	162	226	88
^{90}Rb(158 s)	4135.47(20)	100	100	84	84	84
^{90}Rb(158 s)	4365.82(24)	95	96	100	100	100

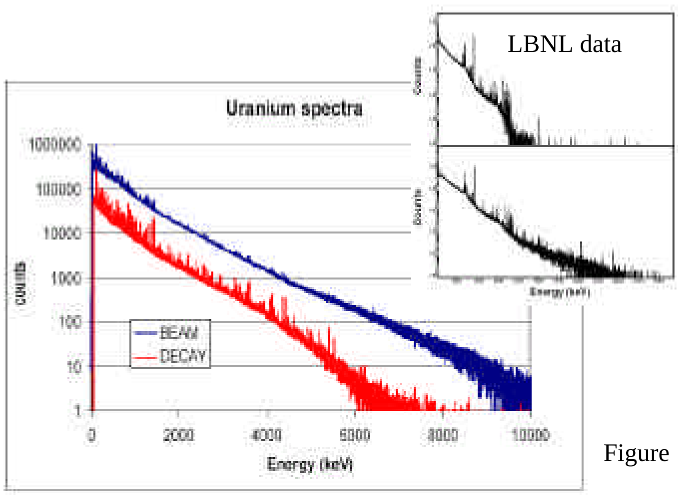

Figure 1. Budapest (^{235}U) and LBNL (^{238}U) Uranium fission data.